

What is claimed is:

1. A process for producing 7-methylxanthine, theobromine or caffeine which comprises: methylation of xanthosine at the 7-position of the purine ring; ribose removal of 7-methylxanthosine at the 9-position of the purine ring; methylation of 7-methylxanthine at the 3-position of the purine ring; and/or methylation of theobromine at the 1-position of the purine ring *ex vivo*, under the catalytic action of a combination of two or more of the following enzymes (a), (c) and (d), and the cellular extract (b):

(a) an enzyme having a catalytic activity of methylation of xanthosine at the 7-position of the purine ring and having the amino acid sequence set out in SEQ ID NO: 1,

(b) a crude cellular extract obtained from *Escherichia coli* having a catalytic activity of ribose removal of 7-methylxanthosine at the 9-position of the purine ring,

(c) an enzyme having a catalytic activity of methylation of 7-methylxanthine at the 3-position of the purine ring and having the amino acid sequence set out in SEQ ID NO: 4,

(d) an enzyme having a catalytic activity of methylation of theobromine at the 1-position of the purine ring and having the amino acid sequence set out in SEQ ID NO: 7.

2. The process according to claim 1 wherein at least

one of the enzymes (a), (c) and (d), and the cellular extract (b) is substituted for one having a comparable activity thereto.

3. A process for producing theobromine or caffeine which comprises allowing the expression of a combination of (a) + (b) or a combination of (a) + (b) + (c) of the following DNA molecules (a), (b) and (c) in a host organism which biologically synthesizes xanthosine and has an enzymatic activity of ribose removal of 7-methylxanthosine to alter the metabolism of the host organism:

(a) a DNA molecule encoding an enzyme which catalyzes methylation of xanthosine at the 7-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 2,

(b) a DNA molecule encoding an enzyme which catalyzes methylation of 7-methylxanthine at the 3-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 5,

(c) a DNA molecule encoding an enzyme which catalyzes methylation of theobromine at the 1-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 8.

4. A process for producing theobromine or caffeine which comprises allowing the expression of a combination of the following DNA molecules (b) and (c) in a host organism which biologically synthesizes 7-methylxanthine to alter the

metabolism of the host organism:

(b) a DNA molecule encoding an enzyme which catalyzes methylation of 7-methylxanthine at the 3-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 5,

(c) a DNA molecule encoding an enzyme which catalyzes methylation of theobromine at the 1-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 8.

5. A process for altering the amount of production of theobromine or caffeine which comprises allowing the expression of a combination of two or more of the following DNA molecules (a), (b) and (c) in a host organism which biologically synthesizes theobromine or caffeine:

(a) a DNA molecule encoding an enzyme which catalyzes methylation of xanthosine at the 7-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO:2,

(b) a DNA molecule encoding an enzyme which catalyzes methylation of 7-methylxanthine at the 3-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 5,

(c) a DNA molecule encoding an enzyme which catalyzes methylation of theobromine at the 1-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 8.

6. The process according to any one of claims 3 to 5 wherein at least one of the DNA molecules (a), (b) and (c) is substituted for one having a comparable function thereto.

7. A process for producing theobromine or caffeine which comprises allowing the expression of a combination of (a) + (b) or a combination of (a) + (b) + (c) of the following RNA molecules (a), (b) and (c) in a host organism which biologically synthesizes xanthosine and has an enzymatic activity of ribose removal of 7-methylxanthosine to alter the metabolism of the host organism:

(a) an RNA molecule encoding an enzyme which catalyzes methylation of xanthosine at the 7-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 3,

(b) an RNA molecule encoding an enzyme which catalyzes methylation of 7-methylxanthine at the 3-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 6,

(c) an RNA molecule encoding an enzyme which catalyzes methylation of theobromine at the 1-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 9.

8. A process for producing theobromine or caffeine which comprises allowing the expression of a combination of the following RNA molecules (b) and (c) in a host organism which

biologically synthesizes 7-methylxanthine to alter the metabolism of the host organism:

(b) an RNA molecule encoding an enzyme which catalyzes methylation of 7-methylxanthine at the 3-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 6,

(c) an RNA molecule encoding an enzyme which catalyzes methylation of theobromine at the 1-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 9.

9. A process for altering the amount of production of theobromine or caffeine which comprises allowing the expression of a combination of two or more of the following RNA molecules (a), (b) and (c) in a host organism which biologically synthesizes theobromine or caffeine:

(a) an RNA molecule encoding an enzyme which catalyzes methylation of xanthosine at the 7-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO:3,

(b) an RNA molecule encoding an enzyme which catalyzes methylation of 7-methylxanthine at the 3-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 6,

(c) an RNA molecule encoding an enzyme which catalyzes methylation of theobromine at the 1-position of the purine ring and having the nucleotide sequence set out in SEQ ID NO: 9.

10. The process according to any one of claims 7 to 9 wherein at least one of the RNA molecules (a), (b) and (c) is substituted for one having a comparable function thereto.

11. The process according to any one of claims 3 to 10 wherein the host organism is a plant, and production or increase of production of theobromine or caffeine defends the host plant from pest feeding by a herbivore.